

Velocity measurements at the metamagnetic transition in UPt_3

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Abstract . The longitudinal ultrasonic attenuation and velocity were measured on a single crystal of UPt_3 in high magnetic fields up to a field of 33 Tesla. Earlier measurements had reported a metamagnetic behavior in the vicinity of 20 Tesla, where a pronounced dip in velocity (softening of the lattice) is seen. Simultaneous measurements at three frequencies during the same run were possible. A small dispersion in the velocity is seen at the metamagnetic transition. An additional structure is seen at a slightly higher magnetic field at lower temperatures. Also at temperatures below 100mK, quantum acoustic oscillations are seen in the ultrasonic velocity.

Keywords . UPt_3 , metamagnetism, velocity dispersion

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1. Introduction

The heavy fermions are rare earth or actinide intermetallic compounds that show strong electronic correlations at low temperatures. This is manifested by an enhanced electronic heat capacity at low temperatures (equivalent to a narrow band at the Fermi level). The renormalized mass may be 100 times larger than the bare-electron mass, leading to the concept of localized electron states, and strong magnetic properties. Some of these are superconducting at temperatures of the order of 1 K, and in these the heat capacity jump at T_c is of the same order as γT showing that the Cooper pairs also involve the heavy mass.

The heavy fermion systems (UPt_3 , UPe_{13} , URu_2Si_2 , UPd_2Al_3 , CeCu_2Si_2 , etc.) have very interesting and unusual superconducting and normal state properties. Ultrasonic velocity methods have proved an important sensitive tool in investigating these properties [1,2]. The best studied of these compounds is UPt_3 . It is now believed that the superconductivity in UPt_3 is of an unconventional nature, *i.e.* not the simple s-wave BCS type. The symmetry of the order parameter is lower than the underlying Fermi surface. There are multiple superconducting phases [2,3] for all orientations of the magnetic field which can be understood in terms of a two component order parameter in the Ginzburg-Landau formalism. A weak antiferromagnetic ordering (at $\sim 5\text{K}$) is instrumental in removing the degeneracy and this shows up

as a splitting in the superconducting phase transition, seen simultaneously by ultrasonic and heat capacity measurements. In addition the ultrasonic attenuation in UPt_3 at low temperatures instead of following an exponential decrease as in a conventional superconductor (because of the development of an energy gap, the number of quasiparticles that attenuate the sound waves drop off exponentially) shows a power law dependence $\alpha \approx T^n$, where $n = 1, 2$, or 3 . For a non-conventional BCS superconductor, the energy gap goes to zero either at points or along lines along the Fermi surface. There is thus always some broken Cooper pairs, and their number has a much weaker temperature dependence giving rise to a power law temperature dependence for the attenuation and the heat capacity. The value of the exponent should tell the difference between a point node or a line node. However the measurements must then be done at very low temperatures, and there seems to be a large variation between samples. Shivaram *et al.* [4] measured the attenuation of shear waves propagating in the basal plane, with the sound polarization vector parallel to and perpendicular to the c-axis. He obtained two different values for n (1 and 2), thereby concluding that the symmetry was that of a *d-wave*.

In strong magnetic fields, some of these compounds (UPt_3 , UAl_2Pd_3 , CeRu_2Si_2 , etc.) show a metamagnetic behavior in which the velocity shows a remarkable dip (softening of the elastic constants) [5]. In UPt_3 , this is about 3% at 1.5K and

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higher at lower temperatures. The metamagnetic transition is at 20.3 T for UPt_3 , 8T for CeRu_2Si_2 and 17.5T for UPd_2Al_3 . These transitions are strongly related to the fact that the heavy fermion state sits close to a magnetic instability. Study of this effect with various probes would throw light on the nature of the heavy fermion state.

2. Experimental methods

Preliminary measurements were done at the High Field Magnet Laboratory at the Max Planck Institute in Grenoble. Recent measurements were done at the National High Magnetic Field Laboratory (NHMFL) at Tallahassee using the 32mm bore 30T and 33T resistive magnets and various cryogenic inserts including a dilution refrigerator that permitted temperature variation between 40mK to 77K. A new pulsed heterodyne ultrasonic spectrometer was built for these measurements, which allowed simultaneous measurement at various frequencies in the same run. Thus there is no ambiguity in the temperature as was in an earlier measurement [6]. The single crystal of UPt_3 was of high quality ($4 \times 4 \times 7\text{mm}$) as evidenced by a sharp superconducting transition ($\sim 15\text{ mK}$) and a relatively high T_c ($\sim 530\text{ mK}$). Overtone-polished LiNbO_3 transducers were affixed with epoxy to the end faces of the crystal. Velocity and attenuation changes were measured using a custom-built pulse superheterodyne phase-sensitive spectrometer. A special feature was the possibility of simultaneously measuring various frequencies, in the same field run. This removed any ambiguity in small temperature variations from one field sweep to another. Relative velocity and attenuation measurements were performed at frequencies 30, 86 and 140 MHz while the magnetic field was swept at a constant rate. The main features were observed with sound propagating in the basal plane, and the field also in the basal plane (UPt_3 is a hexagonal crystal). The easy axis for magnetization is in the basal plane.

3. Experimental results

One of the early signs that ultrasonics is a powerful probe of the heavy fermion state was the observation of an associated peak [7] in the zero field longitudinal sound attenuation near 12K. This attenuation peak can be described in terms of a narrow resonance peak with a width of $\sim 2\text{ meV}$ in the density of states $\sim 1\text{ meV}$ above the fermi surface, presumably resulting from a hybridization of the f levels (or narrow f bands) and the conduction band. A magnetic field of about 20T, comparable to 1 meV in the electronic energy scale, may therefore produce significant effects on the attenuation peak by, *e.g.* shifting and splitting the position of the Fermi surface relative to the resonant peak in the density of states.

The effect of a high magnetic field (up to 23T) on sound velocity and magnetic susceptibility for UPt_3 , was studied by Kouroudis *et al.* [5]; a soft longitudinal acoustic mode and oscillatory behaviors in some shear modes were observed. These observations have been interpreted as a metamagnetic transition

in which the specimen acquires an enhanced magnetic moment within a small field interval: a softening of the lattice, the phenomena observed in acoustic measurements, accompanies this transition. Clearly this is not a true phase transition, in that the thermodynamic properties are not singular (only rapidly varying); it would be worthwhile to carry out the measurements to the lowest possible temperature available, to see if the phenomena evolves into a true phase transition at $T = 0$. With this in mind measurements are being carried out at the National High Magnetic Field Laboratory in Tallahassee on a dilution refrigerator and a 33T magnet.

In an earlier set of measurements at the High Field Magnet Facility at the Max Planck Institute in Grenoble, the velocity and attenuation of sound were measured at fields up to 26T and temperatures down to 2 K. The 'Kondo'-peak seen earlier in the sound attenuation at zero field [7] is enhanced as the magnetic field is increased, and shifts to lower temperatures. However above 17 T, the peak was not observed. The attenuation increased rapidly as the temperature is lowered and the field increased to 20 T. At about 20 T there is a strong velocity dip (elastic softening due to the enhanced magnetization) at the metamagnetic transition with an associated increase in the attenuation. It is interesting to speculate whether the disappearance of the peak in the temperature dependence of the attenuation above 17 T and the onset of the metamagnetic peak in the field dependence at low temperatures may be interpreted as a merging of the two phenomena.

At the metamagnetic transition the longitudinal sound velocity (a measure of the elastic bulk modulus) shows a dip (softening) which becomes larger as the temperature is lowered. This dip is about 1% at 2K and becomes more than 5% at temperatures below 0.5K. There is also a small velocity dispersion (the sound velocity change at the transition is more for sound waves with a lower frequency). Figure 1 shows the large change

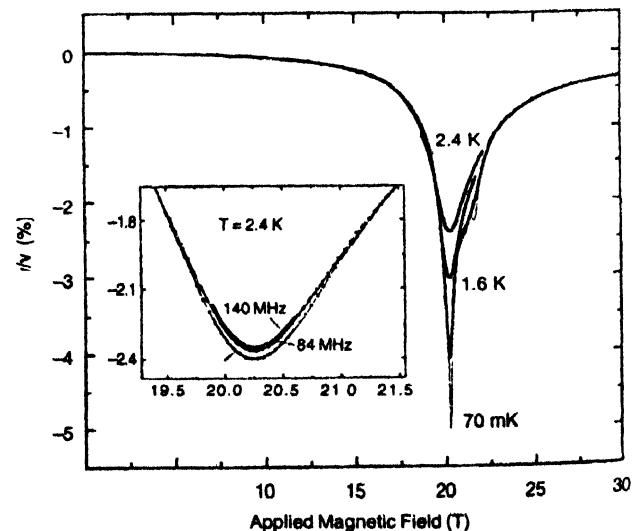


Figure 1. Velocity measurements at the metamagnetic transition at temperatures 2.4K, 1.6K, 0.550K and 0.070K. The inset shows the dispersion for the measurements at 2.4K.

in the velocity at the metamagnetic transition, the velocity dip becoming larger as the temperature is lowered. The inset shows a small velocity dispersion, the measurements being done at the Tallahassee magnet facility. In the earlier measurements at Grenoble, the different frequency runs were done as separate runs, and it was not clear whether the dispersion seen was real, or due to slight variations in the temperature for the different runs. With the new spectrometer, simultaneous measurements were done at the different frequencies, in the same field sweep, so that there is no ambiguity in the temperature. Figure 2 shows the attenuation of the ultrasonic signal as the field is swept. The measurements at different frequencies scale as the square of the frequency. Attenuation measurements could be reliably made only at the lower harmonics.

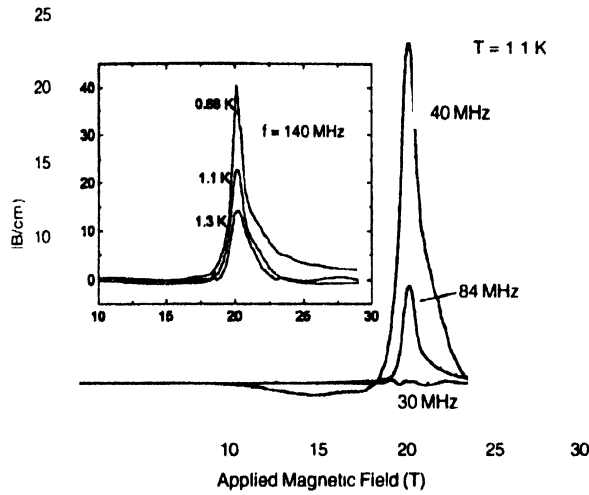


Figure 2. Attenuation at the metamagnetic transition at $T = 1.1\text{ K}$ for frequencies 30, 84 and 140 MHz. The inset shows the temperature dependence of the attenuation measured at 140 MHz.

The magnetization was also measured with a vibrating sample magnetometer. There is an enhanced magnetization at the metamagnetic transition, the feature becoming sharper as the temperature is lowered (see Figure 3). By differentiating, the susceptibility may be obtained: this shows up as a peak at the transition that corresponds to the velocity dip, the two being related thermodynamically through the relation $\Delta c = -\Gamma_H^2 B^2 \chi$, where Δc is the change in the bulk modulus, χ the magnetic susceptibility, B the magnetic field and Γ_H the magnetic Gruenisen parameter. A fit to the data agrees with a value of $\Gamma_H \sim 60$.

At much lower temperature (below 1 K), there seems to be some structure in the velocity dip (a shoulder on the high field side) possibly indicating a second phenomenon (Figure 4). This has now been seen on two different crystals of UPt_3 (grown under different conditions), and there may be a second susceptibility peak at the higher field side of the metamagnetic transition. Susceptibility measurements at lower temperatures are awaited. At still lower temperatures, below $\sim 300\text{ mK}$, Shubnikov-de Haas-like quantum oscillations become visible in

the velocity. To a much lesser extent, oscillatory behaviour is also observed in the attenuation. A detailed analysis of the data is presented in a recent paper [8].

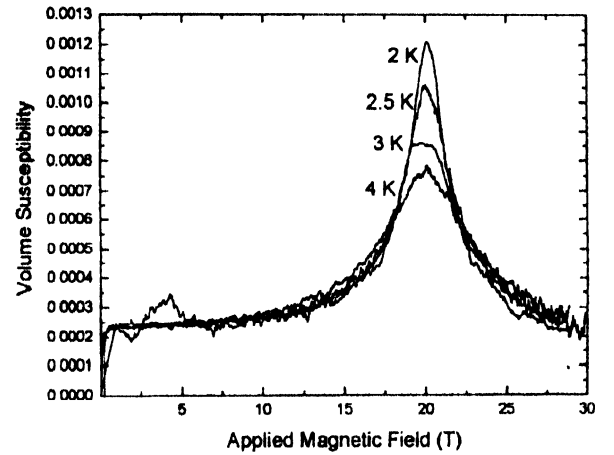
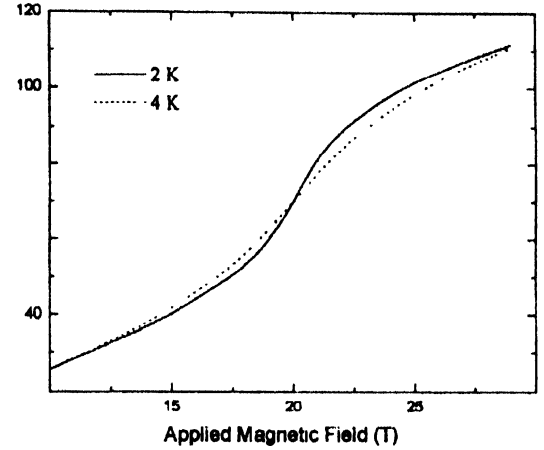


Figure 3. Magnetization measurements (upper curve) on a sample of UPt_3 with a vibrating sample magnetometer. The susceptibility (lower curve) is obtained by differentiating.

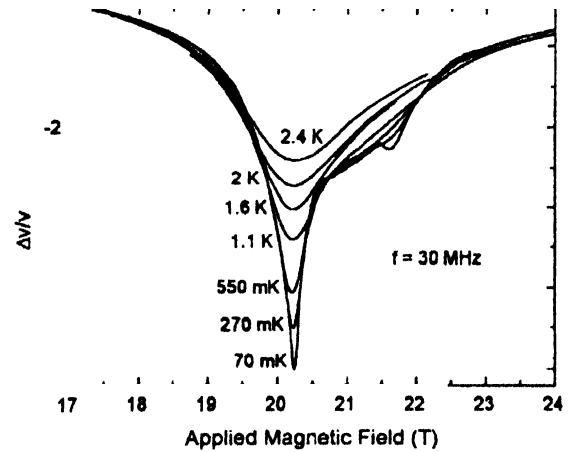


Figure 4. Velocity measurements at the metamagnetic transition at very low temperatures. A second dip is visible at lower temperatures.

4. Conclusion

Ultrasonics has been a powerful probe for the study of phase transitions; both superconducting and magnetic. The velocity

which is a measure of the elastic constants show remarkable features when one goes through either types of phase transition. Most of the striking features were seen with longitudinal sound (the elastic constant involved is the bulk modulus). In the metamagnetic transition, the velocity data agrees with the susceptibility measurements. The added advantage is the much higher sensitivity and the frequency dependence (dispersion). For pure samples and at low temperatures, oscillations in the velocity show that the metamagnetic instability is close to the Fermi surface, and mapping of the Fermi surface may be possible.

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